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| FORM PTO-1390 (Modified) (REV 11-98) | | U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE | | ATTORNEY'S DOCKET NUMBER JMYT-243US | |
| TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371 | | | | U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.5) To Be Assigned 09/857315 | |
| INTERNATIONAL APPLICATION NO. PCT/GB99/03858 | | INTERNATIONAL FILING DATE 22 November 1999 (22.11.99) | | PRIORITY DATE CLAIMED 03 December 1998 (03.12.98) | |
| TITLE OF INVENTION IMPROVEMENTS IN COATINGS | | | | | |
| APPLICANT(S) FOR DO/EO/US de ALVARO, Francisco Lanzacla; MALDE, Chandresh Nemchand; and PETCH, Michael Ian | | | | | |
| Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information: | | | | | |
| 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1). 4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371 (c) (2)) a. <input checked="" type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). b. <input type="checkbox"/> has been transmitted by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)). 7. <input checked="" type="checkbox"/> A copy of the International Search Report (PCT/ISA/210). 8. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). b. <input type="checkbox"/> have been transmitted by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input checked="" type="checkbox"/> have not been made and will not be made. 9. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 10. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). (UNEXECUTED) 11. <input checked="" type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409). 12. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)). Items 13 to 20 below concern document(s) or information included: 13. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 14. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 15. <input checked="" type="checkbox"/> A FIRST preliminary amendment. 16. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. 17. <input type="checkbox"/> A substitute specification. 18. <input type="checkbox"/> A change of power of attorney and/or address letter. 19. <input checked="" type="checkbox"/> Certificate of Mailing by Express Mail 20. <input type="checkbox"/> Other items or information: | | | | | |
| <div style="border: 1px solid black; height: 100px; width: 100%;"></div> | | | | | |

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|---|--|--|--|---|--|
| U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.5) 49/857315 To Be Assigned | | INTERNATIONAL APPLICATION NO. PCT/GB99/03858 | | ATTORNEY'S DOCKET NUMBER JMYT-243US | |
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|---|---------------|--------------|-----------|--|----|-----------------|---------------|---------------|--|
| 21. The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) : <input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$970.00 <input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ⁶ \$840.00 <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$690.00 <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$670.00 <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) \$96.00 ENTER APPROPRIATE BASIC FEE AMOUNT = | | | | CALCULATIONS PTO USE ONLY <table border="1" style="width:100%; border-collapse: collapse;"> <tr> <td style="width:50%;">\$860.00</td> <td style="width:50%;"></td> </tr> <tr> <td>\$0.00</td> <td></td> </tr> </table> | | \$860.00 | | \$0.00 | |
| \$860.00 | | | | | | | | | |
| \$0.00 | | | | | | | | | |
| Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492 (e)). | | | | <table border="1" style="width:100%; border-collapse: collapse;"> <tr> <td style="width:50%;"></td> <td style="width:50%; text-align: right;">\$0.00</td> </tr> </table> | | | \$0.00 | | |
| | \$0.00 | | | | | | | | |
| CLAIMS | NUMBER FILED | NUMBER EXTRA | RATE | | | | | | |
| Total claims | 10 - 20 = | 0 | x \$18.00 | \$0.00 | | | | | |
| Independent claims | 1 - 3 = | 0 | x \$80.00 | \$0.00 | | | | | |
| Multiple Dependent Claims (check if applicable). <input type="checkbox"/> | | | | \$0.00 | | | | | |
| TOTAL OF ABOVE CALCULATIONS = | | | | \$860.00 | | | | | |
| Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) (check if applicable). <input type="checkbox"/> | | | | \$0.00 | | | | | |
| SUBTOTAL = | | | | \$860.00 | | | | | |
| Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492 (f)). | | | | \$0.00 | | | | | |
| TOTAL NATIONAL FEE = | | | | \$860.00 | | | | | |
| Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). <input type="checkbox"/> | | | | \$0.00 | | | | | |
| TOTAL FEES ENCLOSED = | | | | \$860.00 | | | | | |
| | | | | Amount to be: refunded | \$ | | | | |
| | | | | charged | \$ | | | | |

☒ A check in the amount of **\$860.00** to cover the above fees is enclosed.

☐ Please charge my Deposit Account No. _____ in the amount of _____ to cover the above fees.
 A duplicate copy of this sheet is enclosed.

☒ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. **18-0350** A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

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SIGNATURE

Christopher R. Lewis
 NAME

36,201
 REGISTRATION NUMBER

June 4, 2001
 DATE

09/857315

CERTIFICATE OF MAILING BY "EXPRESS MAIL" (37 CFR 1.10)

Applicant(s): Francisco Lanzuela de Alvaro

JCT8 Rec'd PCT/PTO 04 JUN 2001

Docket No.
JMYT-243US

| Serial No. To Be Assigned | Filing Date Herewith | Examiner | Group Art Unit |
|------------------------------|-------------------------|----------|----------------|
|------------------------------|-------------------------|----------|----------------|

Invention: IMPROVEMENTS IN COATINGS

I hereby certify that the following correspondence:

U.S. National Phase Application (w/PTO-1390 and all of the documents listed therein)

(Identify type of correspondence)

is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 in an envelope addressed to: The Assistant Commissioner for Patents, Washington, D.C. 20231

June 4, 2001

(Date)

Kathleen Libby

(Typed or Printed Name of Person Mailing Correspondence)

(Signature of Person Mailing Correspondence)

EL732291839US

("Express Mail" Mailing Label Number)

Note: Each paper must have its own certificate of mailing.

JMYT-243US

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Francisco Lanzuela de Alvaro et al. : Art Unit:
Application No.: To Be Assigned : Examiner:
Filed: Herewith :
FOR: IMPROVEMENTS IN COATINGS :

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
Washington, DC 20231

SIR:

Prior to examination, please amend the above-identified application
as follow.

IN THE CLAIMS:

Please replace claims 1, 3-6, and 8 with the following amended
claims:

1 1. (Amended) A method of depositing metal oxide coatings
2 onto a metal or ceramic substrate comprising the steps of treating a surface of
3 said substrate with a polyelectrolyte to yield a surface coating of the
4 polyelectrolyte thereon, and subsequently or simultaneously treating the
5 surface with an aqueous slurry comprising metal oxide particles.

1 3. (Amended) A method according to claim 1, wherein the
2 metal or ceramic substrate is a catalyst support substrate in plate or monolith
3 form.

1 4. (Amended) A method according to claim 1, wherein the
2 metal oxide is a zeolite.

1 5. (Amended) A method according to claim 1, wherein the
2 metal oxide is selected from alumina, silica, ceria, zirconia, magnesia and
3 mixed oxide catalyst supports.

1 6. (Amended) A method according to claim 4, wherein the
2 metal oxide incorporates a catalytic component.

1 8. (Amended) A supported catalyst comprising a plate or
2 monolith coated with a metal oxide, produced according to claim 1.

Please add the following new claims:

1 9. (Newly Added) A method according to claim 5, wherein
2 the metal oxide incorporates a catalytic component.

1 10. (Newly Added) A method according to claim 9, wherein
2 the catalytic component is one or more of the platinum group metals.

Respectfully submitted,



Christopher R. Lewis, Reg. No. 36,201
Attorney for Applicants

CRL/lrb

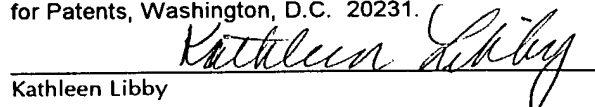
Dated: June 4, 2001

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(610) 407-0700

The Assistant Commissioner for Patents is
hereby authorized to charge payment to
Deposit Account No. 18-0350 of any fees
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EXPRESS MAIL Mailing Label No.: EL732291839US
Date of Deposit: June 4, 2001

I hereby certify that this paper and fee are being
deposited, under 37 C.F.R. § 1.10 and with sufficient
postage, using the "Express Mail Post Office to
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Service on the date indicated above and that the
deposit is addressed to the Assistant Commissioner
for Patents, Washington, D.C. 20231.


Kathleen Libby

VERSION WITH MARKINGS TO SHOW CHANGES MADE

1 1. (Amended) A method of depositing metal oxide coatings
2 onto a metal or ceramic substrate ~~which comprises~~ comprising the steps of
3 treating a surface of said substrate with a polyelectrolyte to yield a surface
4 coating of the polyelectrolyte thereon, and subsequently or simultaneously
5 treating the ~~coated~~ surface with an aqueous slurry comprising metal oxide
6 particles.

1 3. (Amended) A method according to claim 1 ~~or 2~~,
2 wherein the metal or ceramic substrate is a ~~metal or ceramic~~ catalyst support
3 substrate in plate or monolith form.

1 4. (Amended) A method according to claim 1, ~~2 or 3~~,
2 wherein the metal oxide is a zeolite.

1 5. (Amended) A method according to claim 1, ~~2 or 3~~,
2 wherein the metal oxide is selected from alumina, silica, ceria, zirconia,
3 magnesia and mixed oxide catalyst supports.

1 6. (Amended) A method according to claim 4 ~~or 5~~,
2 wherein the metal oxide incorporates a catalytic component.

1 8. (Amended) A supported catalyst comprising a plate or
2 monolith coated with a metal oxide, produced according to ~~any one of the~~
3 preceding claims claim 1.

Claims 9 and 10 have been added.

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09/8573151 GB:009903858

10 Recd. 04 JUN 2001

PFC 1437

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IMPROVEMENTS IN COATINGS

This invention concerns improvements in coatings, and more especially concerns improvements in methods of coating metal oxides and the like onto metallic or non-metallic substrates.

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It is very well established to coat metal oxides, such as alumina, ceria, zirconia, etc, as single or mixed oxides, onto metal substrates and ceramic such as honeycomb through-flow catalyst supports, used in huge numbers in exhaust gas catalysts. This is generally achieved by admixing the metal oxide into an aqueous slurry together with suitable components including adhesion modifiers, of which the "Ludox" (Trade Mark) silica is an example, to form a washcoat slurry. The substrate is then dipped or drenched in the slurry, and fired to yield a high surface area base for the catalyst. The silica may form 50 wt% or more of the solids content of the slurry, and this clearly dilutes the interaction between the catalyst metals and the oxide, which is very important. There has not been significant improvement in these coating techniques for some 20 or 30 years, yet the design of exhaust gas catalysts is becoming more sophisticated and demanding, requiring new components such as zeolites and other multi-layers to be incorporated. We believe that traditional coating methods are failing to deal with new requirements, and in particular, traditional coating methods give problems in multi-layer coating.

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We have previously disclosed the use of polyelectrolytes to coat catalyst particles etc with zeolites formed simultaneously *in situ* (see EP 878 233). We have now discovered that polyelectrolytes can be used to pre-treat metal and ceramic substrates without such *in situ* formation of the zeolites, in a process in which one or more pre-formed metal oxides, including zeolites and the like, is deposited onto the substrate, and not only is a sound, even coating obtained without requiring dilution with adhesion modifiers, (but may include such adhesion modifiers) but the coating is especially suitable for the deposition of further coatings of similar or differing type.

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US 4806427 discloses a process for depositing a catalytically active material onto

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a carrier wherein a polyelectrolyte may be added at 0.2-1.0 percent by weight as an acid-stable liquefier. DE 2300932 discloses coating a polyester resin substrate with a glue which is a polyacrylicamide and a polyvinyl alcohol.

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Accordingly, the present invention provides a method of depositing metal oxide coatings onto a substrate, which comprises treating the surface of said substrate with a polyelectrolyte to yield a surface coating of the polyelectrolyte thereon, and subsequently or simultaneously treating the coated surface with an aqueous slurry comprising metal oxide particles. If required, additional layers of the same or different metal oxide may be applied, either to increase the loading of the metal oxide or to form a multilayer coating.

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Desirably, the polyelectrolyte is applied in the form of an aqueous solution, eg a 0.01 to 20 wt% solution, in the case of the materials specifically described hereinafter a 4 wt%, solution has been found satisfactory. The polyelectrolyte may be any suitable polyelectrolyte, eg anionic or cationic, but polyacrylamides are suitable, such as "Alcostat" (Trademark) 167 available from Allied Colloids Ltd, England. Other polyacrylamides may be considered. For such materials, desirably, the solution is alkaline, for example of pH of approximately 9. Routine testing to optimise the solution pH for each substrate and polyelectrolyte should be undertaken, and may be acid or alkaline, primarily depending upon the surface chemistry of the substrate. Treatment of the substrate may be by any suitable method, including spraying, dipping, vacuum application, drenching using a "waterfall" and the like, and may be carried out at room temperature. Desirably, the polyelectrolyte solution is dried to form a continuous surface polymer layer.

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The substrate may be any metal or ceramic material, in any form such as plate or monolith form, including particularly honeycomb through-flow catalyst supports, but also devices of the type known as static mixers, which provide good gas or other fluid mixing. The metal may be, for example, a stainless steel, including "Fecralloy" or aluminium, and the ceramic may be cordierite or the like. Other substrates which require, or may be protected by, an oxide coating, should also be considered.

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ATT 34/1001

The term metal oxide includes zeolites of all types and of all Si to Al ratios, and includes modified, eg ion-exchanged zeolites, as well as "Silicate" and recently-available zeolite-type compositions where Ga and In or other suitable metals are incorporated into the zeolite framework structure and single or mixed oxides, for example selected from one or more of ceria, zirconia, magnesia, alumina and silica. The metal oxide is desirably in the form of an aqueous slurry, eg of about 40 wt% solids content of particles of the appropriate particle size; if necessary the metal oxide may be milled to disperse the metal oxide whilst simultaneously size reducing. The slurry may, but need not, include an adhesion modifier, and may include other components including catalytically active particles and solutions of catalytically active metals and/or promoters therefor, for example the catalytic component may be one or more of the platinum group metals.

The slurry is then suitably applied by any method to the polyelectrolyte-coated substrate, and is then suitably dried. A final stage is desirably firing, or calcining, to firmly deposit the oxide coating on the substrate. Before the final firing, however, additional slurries of oxide and/or other components may be deposited, to result in a continuous or thicker coating, or a multi-component series of coatings. If desired or necessary, a further polyelectrolyte layer may be applied, and one or more additional metal oxide or other coatings may be applied.

In an alternative embodiment, the polyelectrolyte is admixed with the slurry without a separate pre-coating step. Otherwise the process and materials considerations are fairly similar. This embodiment is expected to be particularly suitable for depositing a thin, even coating.

It is believed, although we do not wish to be bound by any theory, that the polyelectrolyte acts to reverse the charge on the substrate, thus permitting the slurry particles to adhere firmly, and we believe that upon deposition of a second coating, the polyelectrolyte is "re-activated" and serves to cause excellent adhesion of the second coating.

10/11/2000

3a

It is believed that the present invention has significant and unexpected benefits in achieving good coatings on substrates that have previously been difficult to coat, and in particular permits sound and adherent two or multi-layer coatings.

5

A particular embodiment of the invention (according to claim 8) provides a supported catalyst comprising a plate or monolith coated with a metal oxide, produced by the method of the invention.

10

The present invention is illustrated by the following examples.

EXAMPLE 1

15 An aluminium plate is soaked with aqueous 4 wt% polyacrylamide solution at pH 9. The wet plate is then dried to leave a thin surface coating of polyelectrolyte. The coated plate is dipped into a standard zeolite x,y washcoat slurry as used in the

exhaust gas catalyst industry, containing 40 wt% zeolite, with the pH adjusted to between 8 and 9. The washcoat adheres to the plate, and the coated plate is removed and dried at 100 °C for 30 minutes. A further identical washcoat layer was then applied to deposit a desired thickness. The coated metal plate is then calcined at 400°C for 2 hours, which
5 removes the polyelectrolyte residue. A photograph of the thus-coated plate ("A") is shown in the accompanying drawing page. For comparison, the identical procedure was followed without the polyelectrolyte treatment. Although a first coating appeared successful and even, a second coating caused the combined coating to dramatically peel; a photograph of the coated plate is shown as "B" in the drawing. It can readily be seen that the present
10 invention permits an even and continuous coating, whereas double coating without the polyelectrolyte results in flaking of the coating, and uneven, discontinuous coating.

Although the above Example utilised a 100% zeolite coating, other successful coatings can be applied with 50 wt% "Ludox" silica adhesion modifier, or any other
15 proportion.

EXAMPLE 2

Zeolite 13X on Aluminium Sheet

20 An aluminium sheet was coated with zeolite 13X (Zeolyst Int.) using a polyelectrolyte aqueous solution (5% of Allied Colloids BLO6272AQ/Alcostat167, SD n°0004568, and pH 9-10). After having fired the aluminium plate at 200°C, it was anodised in a 15% sulphuric acid solution. The plate was then coated with a thin layer of polyelectrolyte, then four consecutive passes of zeolite 13X were applied with a total
25 weight gain of 0.57g over a surface of approximately 5 in² (32 cm²); this represented a loading improvement over 3 times greater than could be achieved in previous experiments. Between the third and the fourth passes, a second layer of polyelectrolyte was necessary. Finally, the sheet was calcined at 300°C to burn off the polyelectrolyte.

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EXAMPLE 3

Zeolite 13X on Aluminium Monoliths

Two aluminium honeycomb monoliths of different cell density (400 and 200 cells

per sq.in) were coated with zeolite 13X using a polyelectrolyte aqueous solution (5% of Allied Colloids BLO6272AQ/Alcostat167, SD n° 00045768, and pH 9-10). The 400 and 200 cpsi monolith reached coatings of 2.7 and 3.2 g/in³ respectively in fourteen consecutive passes of zeolite 13X with very low percentages of binder (below 5%). A polyelectrolyte layer was applied prior to the first pass and between the 11th and the 13th passes.

EXAMPLE 4

Zeolite Y on Aluminium Sheet

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An aluminium sheet was coated with zeolite Y (Zeolyst Int.) using a polyelectrolyte aqueous solution (5% of Allied Colloids BLO6272AQ/Alcostat167, SD n°00045678, and pH 9-10). The aluminium plate was coated with a thin layer of polyelectrolyte, then three consecutive passes of zeolite Y were applied with a total weight gain of 0.81g over a surface of approximately 1.55in² (10cm²); previous tests using this zeolite without polyelectrolyte showed that the coating would completely disintegrate and peel off after two passes.

EXAMPLE 5

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Zeolite Y on Aluminium Monoliths

An aluminium honeycomb monolith of 200cps cell density was coated with zeolite Y using a polyelectrolyte aqueous solution (5% of Allied Colloids BLO6272AQ/Alcostat167, SD n° 00045768, and pH 9-10). A polyelectrolyte layer was applied prior to the first pass. The monolith reached coatings of 4.2g/in³ of zeolite Y in seven consecutive passes.

EXAMPLE 6

Zeolite 4A on "Fecralloy" static Mixers

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Static mixers (obtainable from Sulzer) made of "Fecralloy" metal were pre-heated to 900°C for 1 hour and soaked in 5% wt Alcostat167 polyelectrolyte solution (Allied Colloids Ltd) at pH 9. The wet mixer is then dried to leave a thin surface polyelectrolyte

coating. The mixer is then dipped in a slurry containing 35wt% zeolite 4A with 5wt% platinum, and 2wt% Ludox silica (DuPont). Excess zeolite is removed by blowing the coated mixer through with 80psi of nitrogen. This helps to free any blocked channels in the mixer. The coated mixer is then dried at 120°C/8hrs, and calcined at 500°C/2hrs. The calcination step burns off the polyelectrolyte layer, and leaves the zeolite coating intact. In this case it results in a thick zeolite layer with coating density of 3.1 ginch⁻³ throughout the mixer.

EXAMPLE 7

Alumina –based Catalyst on “Fecralloy” static Mixers

A static mixer was coated, using the same methodology as in Example 6 with 5% Pt, 0.1%Fe on alumina catalyst. This catalyst was mixed with 7% Dispersal binder (Condea), and made into a 35wt% aqueous washcoat. The resultant coating was observed to have good cohesion and integrity. A coating density of 3.7 ginch⁻³ was achieved.

EXAMPLE 8

Zeolite H-ZSM-5 on Ceramic Monolith

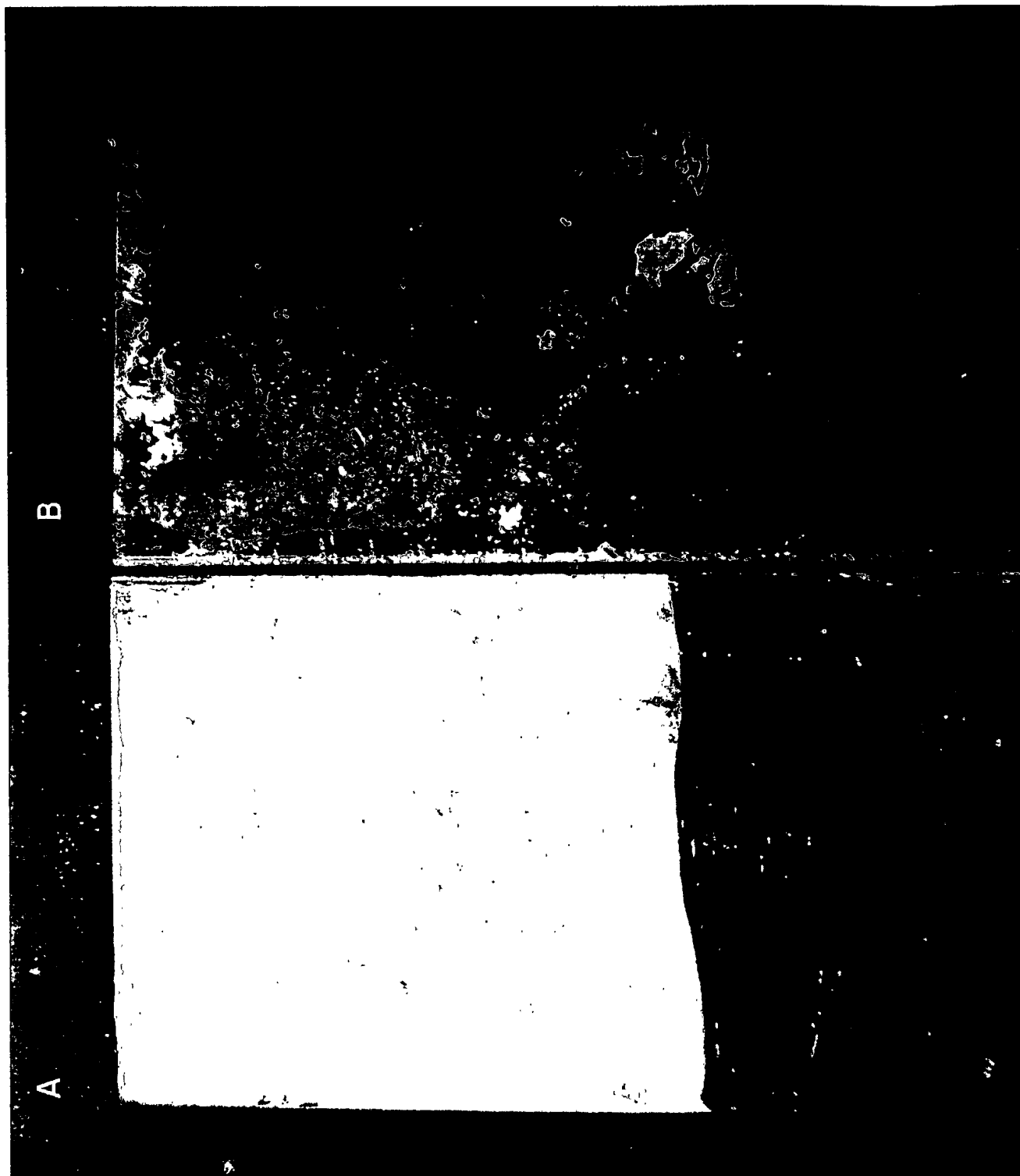
A conventional cordierite monolith was successfully coated with H-ZSM-5 synthetic zeolite using the technique of Example 3. The test was repeated successfully using a mixture of zeolite and colloidal silica binder.

The invention may be modified by the skilled person without departing from the scope thereof.

07-11-2000

CLAIMS

1. A method of depositing metal oxide coatings onto a metal or ceramic substrate
5 which comprises treating a surface of said substrate with a polyelectrolyte to yield a surface coating of the polyelectrolyte thereon, and subsequently or simultaneously treating the coated surface with an aqueous slurry comprising metal oxide particles.
2. A method according to claim 1, wherein said polyelectrolyte is a polyacrylamide.
- 10 3. A method according to claim 1 or 2, wherein the substrate is a metal or ceramic catalyst support substrate in plate or monolith form.
4. A method according to claim 1, 2 or 3, wherein the metal oxide is a zeolite.
- 15 5. A method according to claim 1, 2 or 3, wherein the metal oxide is selected from alumina, silica, ceria, zirconia, magnesia and mixed oxide catalyst supports.
6. A method according to claim 4 or 5, wherein the metal oxide incorporates a
20 catalytic component.
7. A method according to claim 6, wherein the catalytic component is one or more of the platinum group metals.
- 25 8. A supported catalyst comprising a plate or monolith coated with a metal oxide, produced according to any one of the preceding claims.



Declaration and Power of Attorney For Patent Application

English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

IMPROVEMENTS IN COATINGS,

the specification of which is attached hereto unless the following box is checked:



was filed on June 4, 2001 as

United States Application Number or PCT International Application Number 09/857,315

and was amended on June 4, 2001.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)

Priority Not Claimed

9826486.4

Great Britain

03 December 1998

(Number)

(Country)

(Day/Month/Year Filed)

☐

(Number)

(Country)

(Day/Month/Year Filed)

☐

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States provisional application(s) listed below.

(Application Number)

(Filing Date)

(Application Number)

(Filing Date)

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

| | | |
|-------------------------------|------------------------|--|
| _____ (Application Number) | _____ (Filing Date) | _____ (Status - patented, pending, abandoned) |
| _____ (Application Number) | _____ (Filing Date) | _____ (Status - patented, pending, abandoned) |

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

| | | | | | |
|-------------------|-----------------|------------------------|-----------------|-------------------------|-----------------|
| Paul F. Prestia | Reg. No. 23,031 | Lawrence E. Ashery | Reg. No. 34,515 | Jack J. Jankovitz | Reg. No. 42,690 |
| Allan Ratner | Reg. No. 19,717 | Christopher R. Lewis | Reg. No. 36,201 | Jonathan H. Spadt | Reg. No. 45,122 |
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| Kevin R. Casey | Reg. No. 32,117 | Daniel N. Calder | Reg. No. 27,424 | | |
| Benjamin E. Leace | Reg. No. 33,412 | Louis W. Beardell, Jr. | Reg. No. 40,506 | | |
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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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Inventor's signature _____

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Additional inventors are being named on separately numbered sheets attached hereto.

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Third inventor's signature  Date 3rd October 2001

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Full name of fourth joint inventor, if any (given name, family name) _____

Fourth inventor's signature _____ Date _____

Residence _____

Citizenship _____

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Full name of fifth joint inventor, if any (given name, family name) _____

Fifth inventor's signature _____ Date _____

Residence _____

Citizenship _____

Post Office Address _____

Full name of sixth joint inventor, if any (given name, family name) _____

Sixth inventor's signature _____ Date _____

Residence _____

Citizenship _____

Post Office Address _____

Full name of seventh joint inventor, if any (given name, family name) _____

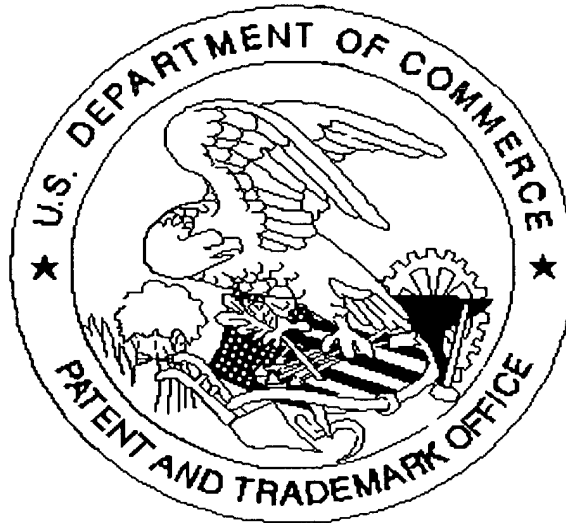
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